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(71) Applicant: MOBIL OIL CORPORATION [US/US]; 3225
Gallows Road, Fairfax, VA 22037-0001 (US).

(72) Inventors: AMBROISE, Benoit; Rue du Moulin, F-55150
Brandeville (FR). BREW, Joseph, Edward; 801 East
Avenue, Newark, NY 14513 (US). KEUNG, Jay, Kin;
6011 Allen Padgham Road, Macedon, NY 14502 (US).
SHEPPARD, Karen, A.; 67 Edendery Circle, Fairport, NY
14450 (US). THE-DZUY, Francis, Tran; 1349 Ayrault
Road, Fairport, NY 14450 (US).

(74) Agents: ROBERTS, Peter, William et al.; Mobil Oil Corpora-
tion, 3225 Gallows Road, Fairfax, VA 22037-0001 (US).

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(54) Title: MULTILAYER ORIENTED HEAT SEALABLE FILM

(57) Abstract

A film structure which includes at least one layer of an olefin homo-, co- or ter-polymer having a surface-treated external surface which is printable, sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-substituted polysiloxane, preferably a cross-linked polymonoalkylsiloxane, and/or liquid polydimethyl siloxane. A method of producing a heat sealable multilayer film structure of improved machinability is also provided.

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MULTILAYER ORIENTED HEAT SEALABLE FILM

This invention relates to the field of composite polymer
5 films of reduced surface frictional characteristics and to a
method of making such films. It more particularly refers to a
biaxially oriented composite film structure having improved
properties, including heat sealability, good machinability and
reduced coefficient of friction.

10 In the packaging of certain types of foods, such as snack
foods including candies, potato chips and cookies, it is common
practice to employ a multi-layer film. Polypropylene films are
widely used in the packaging industry due to their superior
physical properties, such as, transparency, stiffness, moisture
15 barrier characteristics and others. Despite these highly
desirable properties, unmodified polypropylene film has the
disadvantageous property of having a high inherent coefficient
of friction and film-to-film destructive blocking on storage.
This high film-to-film coefficient of friction makes
20 polypropylene films difficult to be successfully employed in
automatic packaging equipment in their unmodified form.

In the past, coefficient of friction characteristics of
polypropylene and other thermoplastic films have been
beneficially modified by the inclusion in the polymer of fatty
25 acid amides, such as, erucamide and oleamide. The effectiveness
of this type of material depends upon its ability to migrate to
the surface of the film in order to reduce the coefficient of
friction. Both of the aforementioned amides are commonly used
for reducing coefficient of friction. The development of the
30 desired low coefficient of friction value is strongly dependent
upon the type and amounts of amides, and time and temperature
aging effects. Even the heat history of the film while in
storage and shipping and during subsequent converter processes,
significantly affects the coefficient of friction. In addition,
35 the presence of these types of fatty acid amides on the film
surface results in visible adverse appearance effects,
manifested by an increase in haze, a decrease in gloss and the

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presence of streaks. These materials also adversely effect the wettability and adhesion of solvent and water-based inks, coatings and adhesives.

In the case of oriented polypropylene films which are widely used in the food packaging industry, it is common to laminate this film with itself or with other thermoplastic films or with paper films. When oleamide or erucamide are used in the polypropylene films a significant increase in coefficient of friction has been observed after lamination to such films. It is theorized that this is due either to the migration of the amide back into the polypropylene film or to the loss of the additive layer at the film surface. Therefore, these types of oriented laminated polypropylene films have limited usage for particular converting processes. Attempts to replace these amides to provide a consistent coefficient of friction have not been successful.

In accordance with the present invention, there is provided a film structure which includes at least one layer of an olefin homo-, co- or ter-polymer having a surface-treated, preferably flame-treated, external surface which is printable, sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-substituted polysiloxane. Particularly preferred particulate cross-linked hydrocarbyl-substituted polysiloxane include the polymonoalkylsiloxanes.

Also, a heat sealable multilayer film structure of improved machinability is provided. The film structure comprises an upper heat sealable layer comprising an olefinic homopolymer copolymer or terpolymer having a surface-treated external surface which is printable, sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-substituted polysiloxane, an intermediate layer comprising an olefinic polymer, and a lower heat sealable layer comprising an olefinic homopolymer, copolymer or terpolymer having an external surface which is sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-

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substituted polysiloxane.

A method of producing a heat sealable multilayer film structure of improved machinability is also provided. The method includes the step of coextruding a film structure, the
5 film structure including an upper heat sealable layer comprising an olefinic homopolymer, copolymer or terpolymer having a surface-treated external surface which is printable, sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-substituted
10 polysiloxane, an intermediate layer comprising an olefinic polymer, and a lower heat sealable layer comprising an olefinic homopolymer, copolymer or terpolymer having an external surface which is sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked
15 hydrocarbyl-substituted polysiloxane. In practicing the method of the present invention, it is particularly preferred to employ as the particulate cross-linked hydrocarbyl-substituted polysiloxane a polymonoalkylsiloxane.

By improved machinability is meant that the film exhibits
20 a consistently low coefficient of friction and has improved anti-slip and non-blocking characteristics.

This invention also provides a composition which comprises (i) a liquid hydrocarbyl-substituted polysiloxane and (ii) a solid particulate cross-linked hydrocarbyl-substituted
25 polysiloxane. The polysiloxanes (i) and (ii) may each be as herein defined. This invention further comprises such a composition which also comprises a thermoplastic polymer matrix. The thermoplastic polymer may be an olefin polymer as herein defined.

30 This invention also relates to the use in a film structure comprising at least one layer of an olefin homo-, co- or terpolymer having a flame-treated external surface, of a particulate cross-linked hydrocarbyl-substituted polysiloxane in a layer to make the surface printable, sealable and
35 machinable.

Accordingly, it is an object of the present invention to provide a film structure of excellent sealability.

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It is another object of the present invention to provide a film structure with improved machinability.

It is a further object of the present invention to provide a film structure having an improved range of process
5 operability.

It is yet another object of the present invention to provide a film which may be bonded to a wide variety of substrates and coatings.

It is a yet further object of the present invention to
10 provide a film structure in which a conventional treatment testing method can be used.

In general, the present invention can be practised with any thermoplastic film, for example, the polyolefins in general, and specifically, polymers, copolymers and blends of ethylene,
15 propylene and butylene, polyesters, such as, polyethylene terephthalate acrylic polymers and copolymers. The basic film structures of the present invention find utility when supported or laminated to another film in order to accommodate a particular end use. Thus, polymer film layers different from
20 the intermediate layer can be laminated to the intermediate layer of the structure for particular purposes. Such polymeric films include any thermoplastic film different from the intermediate layer of the film.

Particularly preferred polymers employed as the
25 intermediate layer of the film herein are the polypropylenes which are highly isotactic. The preferred polypropylenes are well known in the art and are formed by polymerizing propylene in the presence of stereospecific catalysts system. They can have a melt index at 230°C from 0.1 to 25. The crystalline
30 melting point is about 160°C. The number average molecular weight is from 25,000 to 100,000. The density is from 0.90 to 0.91.

For descriptive purpose only, the film structures of the present invention will be described as having an upper skin
35 layer (a), an intermediate layer (b) and a lower skin layer (c). As may be appreciated by those skilled in the art, the use of the terms upper and lower to refer to particular skin layers is

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merely relative. Moreover, although referred to as skin layers, the upper and lower layers may have additional structures bonded thereto, based on the functional requirements of the overall structure.

5 The polymer materials which are contemplated as the material for use in forming skin layers (a) and (c) are suitably exemplified by polyolefinic homopolymers, copolymers and terpolymers. Where homopolymers are to be used, they may be formed by polymerizing the respective monomer. This can be
10 accomplished in a conventional manner by bulk or solution polymerization, as those of ordinary skill in this art will plainly understand. The copolymers are exemplified by and include block copolymers, for example of ethylene and propylene, random copolymers, for example of ethylene and propylene, and
15 other ethylene homopolymers, copolymers, terpolymers, or blends thereof.

Preferred for use in forming upper skin layer (a) and lower skin layer (c) are heat sealable polymeric materials selected from the group consisting of ethylene-propylene-butene-1 (EPB)
20 terpolymer, ethylene-propylene (EP) random copolymer, linear low density polyethylene (LLDPE), low density polyethylene (LDPE), medium density polyethylene (MDPE), high density polyethylene (HDPE) or blends thereof.

Suitable EPB terpolymers are those obtained from the random
25 inter-polymerization of 1 to 8 weight percent ethylene, preferably from 3 to 6 weight percent ethylene with 65 to 95 weight percent propylene, preferably from 86 to 93 weight percent propylene, butene-1 representing the balance. The foregoing EPB terpolymers are for the most part characterized
30 by a melt index at 230°C of 2 to 16 and advantageously from 3 to 7, a crystalline melting point of 100°C to 140°C, an average molecular weight of 25,000 to 100,000 and a density within the range of 0.89 to 0.92 gm/cm³.

The EP random copolymers generally contain from 2 to 7
35 weight percent ethylene, the balance being made up of propylene. The copolymers can have a melt index at 230°C generally from 2 to 15 and preferably from 3 to 8. The crystalline melting

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point is usually from 125°C to 150°C and the number average molecular weight range is from 25,000 to 100,000. The density will usually range from 0.89 to 0.92 gm/cm³.

In general, where blends of EPB terpolymer and EP random copolymer are used, such blends will contain from 10 to 90 weight percent EPB terpolymer and preferably from 40 to 60 weight percent EPB terpolymer, the balance being made up of EP random copolymer.

Heat sealable blends of homopolymer which can be utilized in providing layers (a) and/or (c) include 1 to 99 weight percent polypropylene homopolymer, e.g. one which is the same as, or different from, the polypropylene homopolymer constituting intermediate layer (a) blended with 99 to 1 weight percent of a linear low density polyethylene (LDPE).

Prior to extrusion, in accordance with the present invention, one or both skin layers (a) and/or (c) is compounded with an effective amount of a combined antiblocking and slip agent. Preferred combined antiblocking and slip agents are selected from the group of particulate cross-linked hydrocarbyl-substituted polysiloxanes. Particularly preferred are the particulate cross-linked polymonoalkylsiloxanes. Most particularly preferred are non-melttable polymonoalkylsiloxanes characterized as having a mean particle size of 0.5 to 20.0 μm and a three dimensional structure of siloxane linkages. Such materials are commercially available from Toshiba Silicone Co., Ltd., worldwide, and in the United States from General Electric Co., and are marketed under the tradename Tospearl. Other commercial sources of similar suitable materials are also known to exist.

Although the polymer from which skin layer (a) is formed is not compounded with a silicone oil, this layer may ultimately acquire a machinability-improving and coefficient of friction-reducing amount of silicone oil. Thus, when the finished film laminate containing silicone oil on the exposed surface of skin layer (c) is taken up on a winding coil, some of the silicone oil will be transferred from this surface to the exposed surface of skin layer (a), primarily to the exposed surfaces of the

combined antiblocking and slip agent particles which protrude from layer (a). However, since the interior of layer (a) contains no amount of silicone oil which could interfere with the heat sealing properties of this layer (and ordinarily contains no silicone oil at all), the presence thereon of the transferred silicone oil serves to further enhance the coefficient of friction of the layer, improves its machinability, without significantly impairing its heat sealability.

10 Either or both layers (a) and (c) can also contain pigments, fillers, stabilizers, light protective agents or other suitable modifying ingredients if desired. Further, skin layers (a) and/or (c) can optionally contain a minor amount of an additional antiblock material, such as silica, clays, talc and
15 glass. These antiblock materials can be used alone, or different sizes and shapes can be blended to optimize machinability. The major proportion of these particles, for example, anywhere from more than half to as high as 90 weight percent or more, will be of such a size that a significant
20 portion of their surface area, will extend beyond the exposed surface of such skin layer.

In order to enhance its receptivity for water-based coatings, the upper skin layer can be advantageously treated by flame treating.

25 Lower skin layer (c) of the film laminate can be of substantially the same composition as that of upper surface layer (a). In a preferred embodiment, the polymer constituting layer (c) is compounded with a silicone oil. The silicone oil advantageously possesses a viscosity from 350 to 100,000
30 centistokes with 10,000 to 60,000 centistokes being especially preferred. Examples of suitable silicone oils are polydialkylsiloxanes, polyalkylphenylsiloxanes, olefin-modified siloxane oils, polyether-modified silicone oils, olefin/polyether-modified silicone oils, epoxy-modified silicone oils
35 and alcohol modified silicone oils, polydialkylsiloxanes which preferably have from about 1 to about 4 carbon atoms in the alkyl group, in particular polydimethyl-siloxanes. Of the

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foregoing, the polydialkylsiloxanes, in particular, a polydimethylsiloxane, are preferred for use herein.

The silicone oil is added to layer (c) generally in the form of a dispersion or emulsion, the silicone being present within, as well as on the exposed surface of this layer as discrete microglobules, frequently of an average size from 1 to 2 microns. The silicone oil, which is generally substantially uniformly distributed on the exposed surface of layer (c), is responsible for improving machinability and imparting a further reduction in the coefficient of friction to this surface as well as the exposed surface of layer (a) when some of the oil is transferred thereto after these surfaces have been placed in mutual contact, e.g., as will occur when the laminate film has been wound on a winding coil.

Polydimethylsiloxane or other silicone oils can be present from 0.15 to 1.5 weight percent, preferably less than 0.5 weight percent, of lower layer (c). Some of this silicone oil will, of course, be present on the exposed surface of layer (c). The amount selected should in any event be sufficient to provide a coefficient of friction of layers (a) and (c) (following transfer of silicone oil microglobules to the latter) of about 0.4 or less, preferably between 0.25 to 0.3 up to at least about 60°C. Because of the unique manner in which the silicone oil is applied to just the exposed surface of upper layer (a), such layer exhibits an improved coefficient of friction but not at the expense of its receptivity to water-based coatings, its heat sealability or its optical clarity.

The silicone oil should be incorporated as homogeneously as possible in the polymer constituting layer (c). This can be achieved by either incorporating the silicone oil as a dispersion or emulsion at room temperature and then heating the blend with the application of shearing forces or by incorporating the oil while the blend is being melted. The mixing temperature must be high enough to soften the blend and enable a very uniform mixture to be formed. The temperature required in a kneader or extruder is generally from 170° to 270°C.

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Intermediate layer (b) can also contain anti-static agents, e.g., cocoamine or N,N bis(2-hydroxyethyl) sterylamine. Suitable amines include mono-, di, or tertiary amines.

Intermediate layer (b) will usually represent from 70 to 5 95 percent of the thickness of the overall film laminate or an even higher percentage thereof. Upper skin layer (a) and lower skin layer (c) are coextensively applied to each major surface of intermediate layer (b), usually by being coextruded directly thereon. For example, layers (a), (b) and (c) can be coextruded 10 from a conventional extruder through a flat sheet die, the melt streams being combined in an adapter prior to being extruded from the die. Each of the skin layers (a) and (c) can comprise, for example, approximately 6.0% of the total thickness of the laminate. After leaving the die orifice, the laminate structure 15 is chilled and the quenched sheet then heated and stretched, e.g., five times in the machine direction (MD) and then subsequently, for example, eight times in the transverse direction (TD). The edges of the film can be trimmed. The film laminate is then wound on a reel in order to effect transfer of 20 silicone oil from the exposed surface of layer (c) to the exposed surface of layer (a) as previously explained.

As a result of the biaxial orientation of the film structure herein, several physical properties of the composite layers, such as: flex-crack resistance, Elmendorff tear 25 strength, elongation, tensile strength, impact strength and cold strength properties are improved. The resulting film can have, low water vapor transmission rate characteristics and low oxygen transmission rate characteristics. These improved physical properties make the film ideally suited for packaging food 30 products, even those comprising liquids. The overall thickness of the laminate is not critical and advantageously can be from 0.35 to 2.0 mls.

Where the product of this invention is to be used as a label for goods, whether the label is opaque or transparent is 35 a function of the objectives to be achieved. Where it is desired to hide the contents of the package being labeled, it would be preferable to use an opaque label. However, where it

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is more desirable to expose the contents of the package to customer examination, the label should be transparent.

A suitable peelable label stock is often put up as an assemblage of components. In one such assemblage, the components include: a base liner, a release layer disposed on the base liner, and a label stock, with a suitable pressure sensitive adhesive disposed thereon, releasably adhered to the release layer on the base liner. In instances where the adhesive is a pressure sensitive adhesive, a release layer is disposed on the base liner. In other cases the adhesive may be one activatable by other means, such as, heat and solvent. Where the adhesive is not pressure sensitive, but is activatable by some other means, a release liner is not needed.

As indicated above, instead of a pressure sensitive adhesive, activatable adhesives can be employed for certain labeling techniques. For instance, when a label is to be applied about the full circumference of a package or bottle, water or other solvent can activate an adhesive stripe or strip applied to one end of the label. The label is then fixed in place by a slight overlap of the other end of the label. It has also be discovered that die cutting of labels is materially enhanced by including inorganic particles, such as calcium carbonate in the label skin layer close to the adhesive and/or release layer. These particles may also cause some cavitation as a result.

When an opaque label or film structure is desired, the core or intermediate layer of the film structure of the present invention may be formed in accordance with US 4,377,616.

Where opacifying agents are desired, they may be incorporated in the intermediate composition of this invention, in a proportion of up to about 10%, preferably at least about 1%, by weight. Suitable conventional opacifying agents can be added to the melt mixture of the intermediate polymer before extrusion thereof into a film. Opacifying compounds are generally well known in this area. They may be exemplified by iron oxides, carbon black, aluminum, aluminum oxide, titanium dioxide, and talc.

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The processability and machinability of the film may be further enhanced by the inclusion in the polymeric material used to form one or both skin layers of a small percentage of finely subdivided inorganic material. Such inorganic material not only can impart antiblock characteristics to the multi-layer film structure of the present invention, but also can reduce the coefficient of friction of the resultant film.

Contemplated finely divided inorganic materials, referred to above may be exemplified by: syloid, a synthetic amorphous silica gel, having a composition of about 99.7% SiO₂; diatomaceous earth having a composition of, for example, 92% SiO₂, 3.3% Al₂O₃, and 1.2% Fe₂O₃ which has an average particle size of about 5.5 microns, which particles are porous and irregularly shaped; dehydrated kaolinite (Kaopolite SF) having a composition of 55% SiO₂, 44% Al₂O₃ and 0.14% Fe₂O₃, which has an average particle size of about 0.7 microns, and which particles are thin flat platelets; and synthetic, precipitated silicates, for example Sipernat 44, a material having a composition of 42% SiO₂, 36% Al₂O₃, and 22% Na₂O, which has an average particle size of about 3-4 microns, and in which the particles are porous and irregularly shaped.

The polyolefin blends used to coextrude the multi-layer high opacity film structures contemplated herein are suitably formed by employing commercially available intensive mixers, such as those of the Bolling or Banbury type.

If desired, the exposed surface of skin layers (a) and/or (c) may have applied to it, coating compositions or substrates such as another polymer film or a laminate; a metal foil, such as aluminum foil; cellulosic webs, e.g. numerous varieties of paper, such as corrugated paperboard, craft paper, glassine, cartonboard, nonwoven tissue, e.g. spunbonded polyolefin fiber and melt-blown microfibers. The application may employ a suitable adhesive, e.g. a hot melt adhesive, such as low density polyethylene, ethylene-methacrylate copolymer; a water-based adhesive such as polyvinylidene chloride latex.

The following specific Examples illustrate this invention. Unless indicated to be on some other basis, all parts and

percentages are by weight.

Coefficient of friction values referred to herein have been determined according to the procedure of ASTM D 1894-78. Haze and gloss values referred to herein were determined according to the procedures of ASTM D 1003-61 and D 2457-70, respectively.

In the following comparative examples, Examples 1-3, Example 1 illustrates a film laminate containing erucamide, a fatty acid amide slip agent, and as such, is representative of a known type of composite film. Example 2 demonstrates the heat sealability-impairing effect of a laminate in which silicone oil has been compounded in the polymers constituting both layers (a) and (c). Example 3 is illustrative of a laminate film and film forming method wherein silicone oil is compounded in the polymer constituting layer (c).

15

EXAMPLE 1

A intermediate layer (b) of about 20 microns thickness derived from a propylene homopolymer of high stereoregularity containing 750 ppm erucamide is melted and coextruded with an upper skin layer (a) of about 0.61 microns thickness derived from an ethylene-propylene-butene-1 terpolymer or ethylene-propylene random copolymer and a lower skin layer (c) of about 1.2 microns thickness of the aforesaid ethylene-propylene-butene-1 terpolymer. The EPB terpolymer components of layers (a) and (c) each contains about 0.2 weight percent of synthetic amorphous silica in the form of approximately spherical particles averaging 3 microns diameter. The extrudate is then cooled, reheated and stretched biaxially 4-6 times in the machine direction and 8-10 times in the transverse direction. Subsequently, upper skin layer (a) is corona treated in a conventional manner and wound in a mill roll form. After 1-3 days of hot room aging at 100°-125°F, the coefficient of friction (COF) of layers (a) and (c) is 0.26 and 0.29 respectively.

35

However, it must be noted that since the film of this example utilizes a migratory slip agent which requires hot room aging, its COF value is often inconsistent and unpredictable.

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Also, as shown in the table below, layer (a) was not receptive to a water-based polyvinylidene chloride adhesive.

EXAMPLE 2

5 Example 1 was substantially repeated but using an isotactic polypropylene as the resin constituting intermediate layer (b). Erucamide was not present. Polydimethylsiloxane at 0.6 weight percent was compounded in layers (a) and (c). As shown from the data in the accompanying table, coefficient of friction behavior
10 was acceptable and water-based ink and water-based adhesive wet-out/adhesion were acceptable. However, the heat sealability capability of layer (a) was essentially destroyed due to the presence of the silicone oil in layer (a).

15

EXAMPLE 3

Example 2 was substantially repeated but with 0.6 weight percent of polydimethylsiloxane compounded in layer (c) only. Layer (a) contained no silicone oil. The improvement of coefficient of friction equilibrium, good converting
20 characteristics of the PVDC adhesive and retention of heat sealability are noted from the data in the accompanying table. The resultant film also possesses good optical clarity characteristics. However, this film will not machine adequately on the horizontal or vertical form fill and seal machines.

25 Film properties are compared for the films of Examples 1-3 in Table 1, below.

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TABLE 1

| Example | Treated Side | Slip | Treated To Treated COF | Untreated To Untreated COF | Crimp Seal Strength 240°F 3/4 Sec. Treated/Treated 425 gm/in |
|---------|--------------|------------------------------------|------------------------|----------------------------|--|
| 1 | Layer (a) | 750 ppm Erucamide | .26 | .22 | 0 |
| 2 | Layer (a) | Silicone oil in Layers (a) and (c) | .30 | .30 | 0 |
| 3 | Layer (a) | Silicone oil in Layer (c) only | .30 | .30 | 475 gm/in |

EXAMPLE 4

25 The coextruded biaxially oriented film structure of this example consists of a polypropylene intermediate layer with two outer heat sealant layers containing the particulate cross-linked polymonoalkylsiloxane non-migratory combined antiblock and slip agent (GE Tospearl RS-344). Two

30 terpolymer sealant resins were used, each containing a loading of 2500 ppm of the non-migratory slip agent, with a particle size of 4.5 microns. The total gauge of this particular film structure was 1.25 mil, with target skin thicknesses of 2.5 gauge (upper) and 4.0 gauge (lower). The

35 film structure was biaxially oriented and flame treated on one side to improve its wettability and to optimize the printability and lamination strengths.

The resultant biaxially oriented film structure had the following properties tested immediately off-line:

40

- 15 -

TABLE 2

| 5 | Minimum Seal Temperature | | Coefficient of Friction | | <u>Haze</u> |
|--------------|--------------------------|---------------|-------------------------|---------------|-------------|
| | <u>T/T(1)</u> | <u>U/U(2)</u> | <u>T/T(1)</u> | <u>U/U(2)</u> | |
| <u>Gloss</u> | 227 | 216 | 0.11 | 0.14 | 2.1 |
| 86.5 | | | | | |

(1) Treated surface to treated surface.

10 (2) Untreated surface to untreated surface.

This film structure had good COF properties, however, its machinability was poor.

15

EXAMPLE 5

The film structure of Example 5 had an identical resin as the intermediate (b) layer as Example 4, but utilized different skin layer (a) and skin layer (c) resins. The upper skin layer (a) had 2000 ppm of the non-migratory
 20 antiblock, and the lower skin layer (c) had 1000 ppm, both 4.5 micron size (GE Tospearl RS-344). The lower skin layer (c) also had the addition of 4000 ppm silicone oil. The film structure was flame treated on one side to improve its wettability and to optimize the printability and lamination
 25 strengths.

The resultant biaxially oriented film structure had the following properties tested immediately off-line:

TABLE 3

| 30 | Minimum Seal Temperature | | Coefficient of Friction | | <u>Haze</u> |
|--------------|--------------------------|---------------|-------------------------|---------------|-------------|
| | <u>T/T(1)</u> | <u>U/U(2)</u> | <u>T/T(1)</u> | <u>U/U(2)</u> | |
| <u>Gloss</u> | 240 | 226 | 0.13 | 0.14 | 1.7 |
| 35 85.3 | | | | | |

(1) Treated surface to treated surface.

(2) Untreated surface to untreated surface.

40

This film structure had good COF and hot slip properties, and also machined well.

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EXAMPLE 6

The film structure of Example 6 was identical to the Example 5 structure, except for slightly lower non-migratory antiblock loading levels (again, GE Tospearl RS-344 was used) and the addition of 1000 ppm silica antiblock agent, 4 microns in size, to both the upper and lower skin layers (a and c). The film structure was flame treated on one side to improve its wettability and to optimize the printability and lamination strengths.

10 The resultant biaxially oriented film structure had the following properties tested immediately off-line:

TABLE 4

| 15 | Minimum Seal Temperature | | Coefficient of Friction | | Haze |
|----|--------------------------|---------------|-------------------------|---------------|------|
| | <u>T/T(1)</u> | <u>U/U(2)</u> | <u>T/T(1)</u> | <u>U/U(2)</u> | |
| | <u>Gloss</u> | | | | |
| | 225 | 234 | 0.31 | 0.15 | 1.2 |
| | 88.1 | | | | |

20

- (1) Treated surface to treated surface.
 (2) Untreated surface to untreated surface.

25

The film structure of this example had good COF and improved hot slip properties. The machinability was also found to be very good.

EXAMPLE 7

30

This example utilizes a high density polyethylene as the upper skin layer (a), which contains 1600 ppm of particulate cross-linked polymonoalkylsiloxane non-migratory combined antiblock and slip agent (again, GE Tospearl RS-344 was used) and a blend of low density polyethylene. The intermediate layer is identical to that described in Examples 4, 5 and 6. The lower skin layer (c) resin contained 1600 ppm of the same non-migratory combined antiblock and slip agent. The overall gauge of this film structure was 0.7 mil,

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with target skin thicknesses of 3 gauge for the upper skin layer and 4 gauge for the lower skin layer. The film was flame treated on one side to improve its wettability and to optimize the printability and lamination strengths.

5 The resultant biaxially oriented film structure had the following properties tested immediately off-line:

TABLE 5

| Minimum Seal Temperature | | Coefficient of Friction | | Haze |
|--------------------------|---------------|-------------------------|---------------|------|
| <u>T/T(1)</u> | <u>U/U(2)</u> | <u>T/T(1)</u> | <u>U/U(2)</u> | |
| 232 | 219 | 0.21 | 0.65 | 1.5 |
| <u>Gloss</u> | | | | |
| 85.5 | | | | |

- 15 (1) Treated surface to treated surface.
 (2) Untreated surface to untreated surface.

This film had good COF properties, on the treated side, and marginal machinability.

20 The table below summarizes the properties of each of the examples illustrated above:

TABLE 6

| Example No. | Minimum Seal Temperature | | Coefficient of | |
|-------------|--------------------------|---------------|----------------|---------------|
| | <u>T/T(1)</u> | <u>U/U(2)</u> | <u>T/T(1)</u> | <u>U/U(2)</u> |
| 4 | 227 | 216 | 0.11 | 0.14 |
| 2.1 | 86.5 | | Poor | |
| 5 | 240 | 226 | 0.13 | 0.14 |
| 85.3 | Good | | | 1.7 |
| 6 | 225 | 234 | 0.31 | 0.15 |
| 88.1 | Good | | | 1.2 |
| 7 | 232 | 219 | 0.21 | 0.65 |
| 85.5 | Marginal | | | 1.5 |

- (1) Treated surface to treated surface.
 (2) Untreated surface to untreated surface.
 (3) Machinability indicates the film's performance on both horizontal and vertical packaging machines.

CLAIMS:

1. A heat sealable multilayer film structure of improved machinability which comprises
- 5 (a) an upper heat sealable layer comprising an olefinic homopolymer, copolymer or terpolymer having a surface-treated external surface which is printable, sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-
- 10 substituted polysiloxane;
- (b) an intermediate layer comprising an olefinic polymer; and
- (c) a lower heat sealable layer comprising an olefinic homopolymer, copolymer or terpolymer having an
- 15 external surface which is sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-substituted polysiloxane.
- 20 2. A multilayer film structure according to claim 1, wherein layer (c) further comprises a liquid hydrocarbyl-substituted polysiloxane.
3. A multilayer film structure according to claim
- 25 1 or 2, wherein the particulate cross-linked hydrocarbyl-substituted polysiloxane of layer (a) or (c) is a cross-linked polymonoalkylsiloxane.
4. A multilayer film structure according to any
- 30 preceding claim, wherein the particulate cross-linked hydrocarbyl-substituted polysiloxane has a mean particle size from 0.5 to 20.0 μm .
5. A multilayer film structure according to any
- 35 preceding claim, wherein at least one outer surface thereof is treated by flame treating.

- 19 -

6. A composition which comprises (i) a liquid hydrocarbyl-substituted polysiloxane and (ii) a solid particulate cross-linked hydrocarbyl-substituted polysiloxane.

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7. A composition according to claim 6 wherein (ii) comprises a cross-linked polymonoalkylsiloxane.

8. A composition according to claim 6 or 7
10 wherein (ii) has a mean particle size from 0.5 to 20.0 μ m.

9. A composition according to claim 6, 7 or 8 which also comprises a thermoplastic polymer matrix.

15 10. A composition according to claim 9 wherein the thermoplastic polymer comprises an olefin homo-, co- or ter-polymer.

20 11. A film structure comprising at least one layer of an olefin homo-, co- or ter-polymer having a flame-treated external surface which is printable, sealable and machinable, the layer containing, as combined slip agent and antiblock, a particulate cross-linked hydrocarbyl-substituted polysiloxane.

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12. A film structure according to claim 1 wherein the external surface also comprises a liquid hydrocarbyl-substituted polysiloxane.

30 13. Use, in a film structure comprising at least one layer of an olefin homo-, co- or ter-polymer having a flame-treated external surface, of a particulate cross-linked hydrocarbyl-substituted polysiloxane in a layer to make the surface printable, sealable and machinable.

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US94/14280

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : B32B 5/16, 27/02, 27/08, 27/32; C08F 283/00; C08K 5/54
US CL : 428/323, 331, 405, 515, 516, 523; 525/477; 524/267

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/323, 331, 405, 515, 516, 523; 525/477; 524/267

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|-----------|--|-----------------------|
| Y | US, A, 4,966,933 (KAWAKAMI ET AL) 30 October 1990, column 2, lines 38-40; column 5, lines 1-25; column 3, lines 37-51. | 1, 3, 8, 11, 13 |
| Y | US, A, 5,110,671 (BALLONI ET AL) 05 May 1992, Abstract; column 3, lines 1-13 and 46-60. | 1, 2, 6, 11- 13 |
| Y | US, A, 4,761,327 (HAMANO ET AL) 02 August 1988, column 4, lines 40-68. | 3, 7 |

Further documents are listed in the continuation of Box C. See patent family annex.

| | |
|---|--|
| * Special categories of cited documents: | *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention |
| *A* document defining the general state of the art which is not considered to be part of particular relevance | *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone |
| *E* earlier document published on or after the international filing date | *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art |
| *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) | *B* document member of the same patent family |
| *O* document referring to an oral disclosure, use, exhibition or other means | |
| *P* document published prior to the international filing date but later than the priority date claimed | |

Date of the actual completion of the international search

27 JANUARY 1995

Date of mailing of the international search report

23 FEB 1995

Name and mailing address of the ISA/US
Commissioner of Patents and Trademarks
Box PCT
Washington, D.C. 20231

Authorized officer

Paul Thibodeau
PAUL THIBODEAU

Facsimile No. (703) 305-3230

Telephone No. (703) 308-2351

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US94/14280

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. Claims Nos.: 4, 5, 9 & 10
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- The additional search fees were accompanied by the applicant's protest.
 No protest accompanied the payment of additional search fees.